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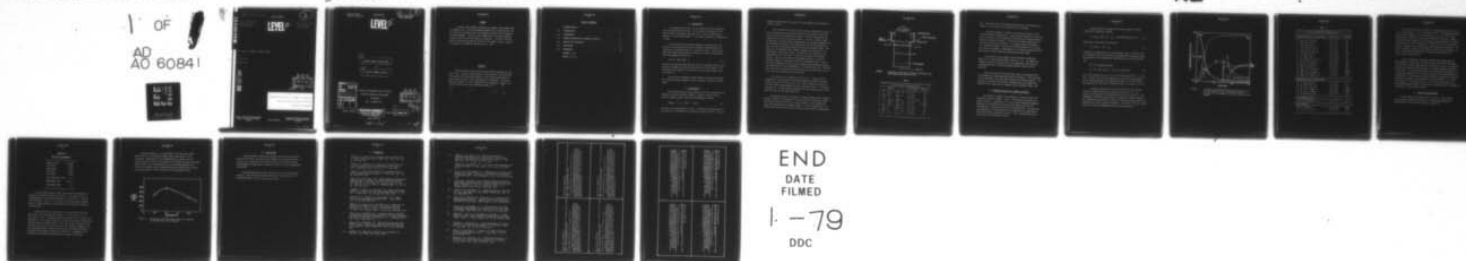
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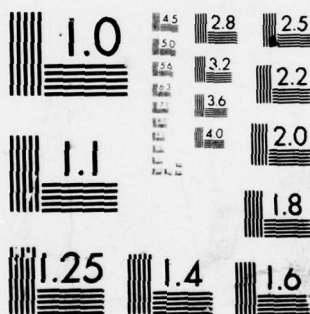
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A PURELY CHEMICAL HBr LASER

S.J. Arneld

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RESUME

L'action laser purement chimique avec du HBr a été obtenue pour la première fois. Du chlore atomique produit par l'interaction du NO et du ClO_2 a été combiné avec des molécules de brome afin de produire du brome atomique. Celui-ci a été combiné avec du HI pour produire du HBr excité sur des transitions de vibration. Le laser a une puissance de sortie multiligne de 0.58 W sur les lignes $P_{1 \rightarrow 0}(4)$, $P_{1 \rightarrow 0}(5)$ et $P_{1 \rightarrow 0}(6)$ de la bande de vibration $v=1 \rightarrow 0$. (NC)

ABSTRACT

Purely chemical laser action in HBr has been achieved for the first time. Chlorine atoms produced by the reaction of NO with ClO_2 were reacted with molecular bromine to form bromine atoms which in turn reacted with HI to form vibrationally excited HBr. Multiline HBr laser output of 0.58 W originated from the $P_{1 \rightarrow 0}(4)$, $P_{1 \rightarrow 0}(5)$ and $P_{1 \rightarrow 0}(6)$ lines of the $v=1 \rightarrow 0$ vibrational band. (U)



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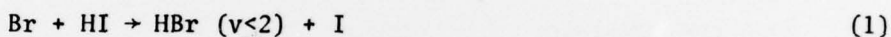
FIGURES 1 to 3

TABLES I to III

1.0 INTRODUCTION

In recent publications [1,2] we reported the first operation of a purely chemical HCl laser. We now report the operation of a purely chemical continuous wave HBr laser. Both of these lasers operate in a wavelength region ($\sim 4 \mu\text{m}$) where atmospheric transmission is high.

As in the case of the purely chemical HCl laser [1,2], the reaction of NO with ClO_2 was used to generate nonequilibrium concentrations of chlorine atoms in a fast flow system. The chlorine atoms then reacted with molecular bromine to produce bromine atoms. The laser pumping reaction was

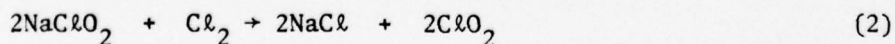


Chen [3] has reported laser emission from this pumping reaction when molecular bromine (Br_2) was flash photolysed in the presence of HI. Transitions from both the $v=1 \rightarrow 0$ and $v=2 \rightarrow 1$ vibrational bands of HBr were observed.

This work was performed at DREV between October and December, 1977 under PCN 33H07 (formerly 21T02), Research on Chemical Excited Lasers.

2.0 EXPERIMENTAL

The chlorine dioxide (ClO_2) required for these experiments was prepared by passing chlorine through a column containing sodium chlorite (NaClO_2) flakes where the heterogeneous reaction



occurred. The concentration of ClO_2 is determined by measurement of the intensity of absorption of ultraviolet light at 3515 \AA . The pro-

cedure for generating $\text{C}\ell\text{O}_2$ and for its measurement are described in detail in Ref. 2.

The transverse flow laser used is shown schematically in Fig. 1. The laser body was constructed of stainless steel and its interior metal surfaces were covered with Teflon. The flow channel was rectangular with a cross-sectional area of 1×40 cm. A choke screen consisting of a Teflon gasket through which 243 uniformly spaced holes were drilled was placed at the upstream end of the flow channel to make the flow of $\text{C}\ell\text{O}_2/\text{He}$ uniform across the full channel width. The first row of stainless steel tubular injectors was located 6.4 cm downstream of the choke screen. A second row was located 11.4 cm downstream of the first one, in line with the edge of the window section. Each row consisted of 60 tubular injectors, 0.85 mm in internal diameter, containing eleven 0.3-mm-diameter holes. These holes were disposed so that the gas passing through them entered near the center plane of and perpendicular to the mainstream flow. With the first injector row used for NO injection and the second for HI injection, this device operated as an HCl laser.

For the device to operate as an HBr laser, a third injector assembly for the injection of molecular bromine was added 9 cm downstream of the first injector row. This third injector consisted of a 1/4-inch Teflon tube flattened into an elliptical shape. It contained 120 holes of ~ 0.3 -mm diameter oriented such that injection was in the same direction as the mainstream flow.

The optical cavity consisted of a protected metal total reflector having a radius of curvature of 10 m and a partially reflecting (98%) multilayer dielectric mirror on a CaF_2 substrate. The windows were CaF_2 flats mounted at the Brewster angle. A flow of helium near the windows kept the surface clean and the optical path free of deactivated

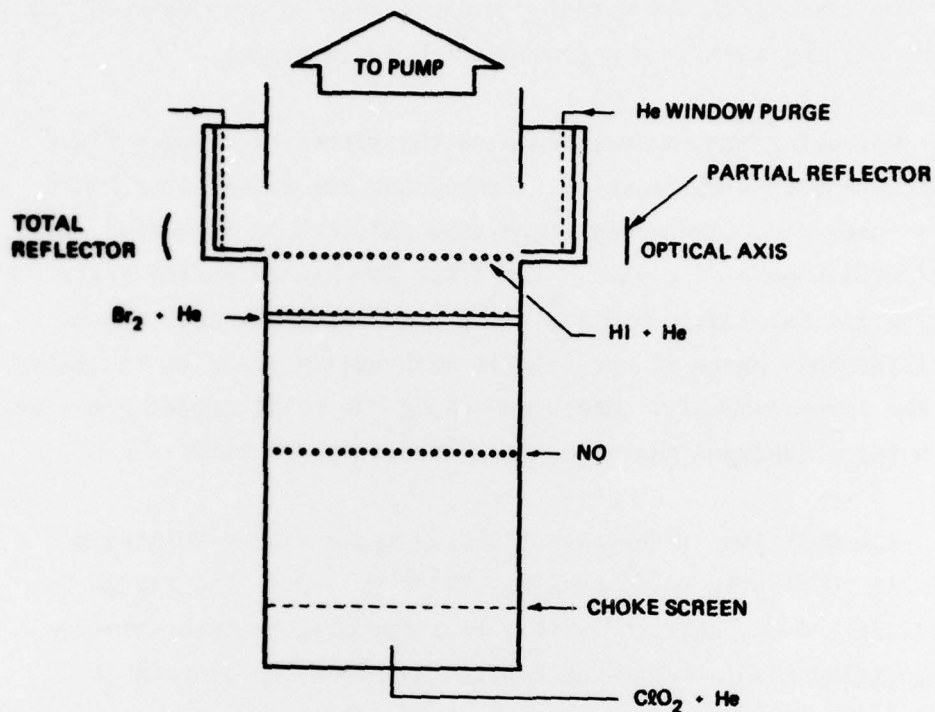


FIGURE 1 - Schematic of HBr purely chemical continuous wave laser employing transverse flow

TABLE I

Principal reactions and their rate constants for the $\text{NO}-\text{ClO}_2-\text{Br}_2-\text{HI}$ system.
All rate constants are in molecular units

Process	Rate coefficient	Reference
1. $\text{NO} + \text{ClO}_2 \rightarrow \text{NO}_2 + \text{ClO}$	3.4×10^{-13}	[7]
2. $\text{NO} + \text{ClO} \rightarrow \text{NO}_2 + \text{Cl}$	1.7×10^{-11}	[8]
3. $\text{ClO}_2 + \text{Cl} \rightarrow 2\text{ClO}$	5.9×10^{-11}	[7]
4. $\text{Cl} + \text{NO}_2 + \text{M} \rightarrow \text{NO}_2\text{Cl} + \text{M}$	7.2×10^{-31}	M=Ar [15]
5. $\text{Cl} + \text{NO}_2\text{Cl} \rightarrow \text{NO}_2 + \text{Cl}_2$	3.0×10^{-11}	estimated
6. $\text{NO}_2 + \text{ClO} + \text{M} \rightarrow \text{NO}_3\text{Cl} + \text{M}$	1.0×10^{-31}	[16]
7. $\text{Br}_2 + \text{Cl} \rightarrow \text{BrCl} + \text{Br}$	1.2×10^{-10}	[4]
8. $\text{BrCl} + \text{Cl} \rightarrow \text{Br} + \text{Cl}_2$	1.43×10^{-11}	[4]
9. $\text{Br} + \text{HI} \rightarrow \text{HBr} + \text{I}$	1.0×10^{-11}	[5]

HBr. The mirrors were externally mounted and the area between the windows and the mirrors was flushed with dry nitrogen.

Molecular bromine was added to the system by using a fluid injector-vaporizer combination. Liquid bromine passed at a known rate from a reservoir, down a capillary tube and into an evacuated flask, filled with chunks of copper. The flask was heated sufficiently to vaporize all the liquid bromine. The bromine vapour then passed to a large bulb where it was diluted with helium prior to its entry into the laser assembly. Measurements of the total output power were made using a Coherent Radiation Laboratories power meter.

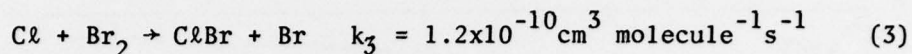
The multiline laser output was analyzed with a McPherson Model 218 0.3-meter monochromator blazed at 3 μm . The signal was chopped at 200 Hz, detected with a room temperature InSb detector and amplified with a Princeton Applied Research (HR-8) Lock-In Amplifier.

Molar flow rates of the individual gases were measured with either Matheson or Hastings-Raydist mass flow meters. Back pressures in the individual flow lines were measured with Helicoïd gauges. The pressure in the main flow channel containing the laser region was measured with a quartz spiral gauged (Texas Instruments Model 145).

3.0 COMPUTER SIMULATION OF CHEMICAL KINETICS

The kinetics leading to the formation of chlorine atoms were discussed in detail in Refs. 1 and 2. The principal reactions are given in Table I. Reactions 1 to 3 form a chain-branching mechanism leading to the formation of chlorine atoms when the initial NO concentration is twice the initial $\text{C}\ell\text{O}_2$ concentration. The principal loss mechanisms for $\text{C}\ell\text{O}$ radicals and $\text{C}\ell$ atoms are the termolecular reactions with NO_2 (reactions 6 and 4). For $\text{C}\ell$ atoms the NO_2 acts catalytically via reactions 4 and 5.

Atomic bromine is formed by the rapid reaction of atomic chlorine with molecular bromine.

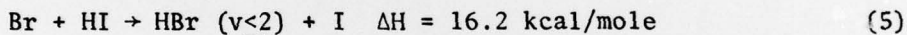


which may be followed by the reaction



leading to the formation of a second bromine atom. As was the case for chlorine atoms, the principal loss mechanisms for bromine atoms are expected to be termolecular recombinations, possibly involving NO_2 .

The laser pumping reaction



has a rate constant of $(1.0 \pm 0.3) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ at 300 K [5]. Polanyi et al [6] have shown that 50-60% of the exothermicity of the reaction appears as vibrational energy in the newly formed HBr bond and that the ratio of formation into $v=1$ and $v=2$ is 8 to 10.

A computer simulation of the time evolution of the various species in the $\text{NO-ClO}_2\text{-Br}_2\text{-HI}$ system is given in Fig. 2. The reactions considered in this simulation are given in Table II. The assumption of instantaneous mixing of reactants was made throughout the computations since kinetic trends and processes, rather than fluid dynamic ones, were of primary importance. The effect of finite mixing time is to limit the formation rates of certain species.

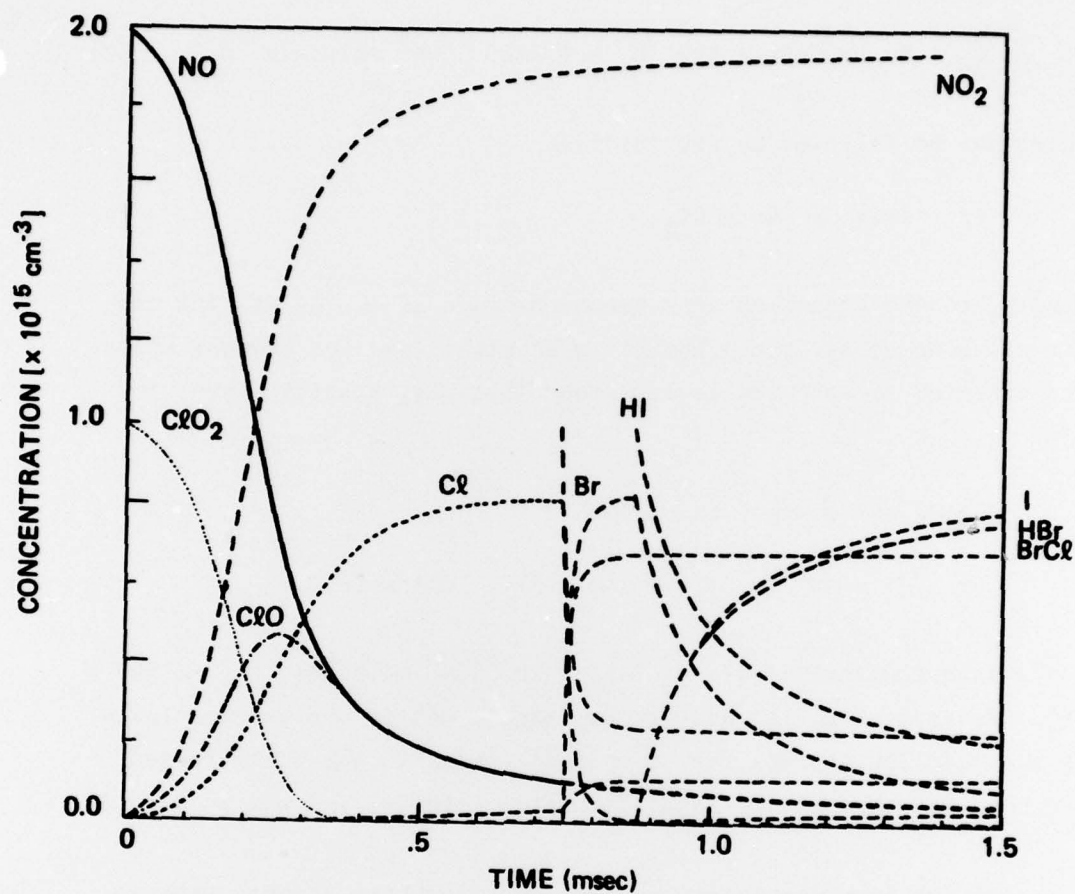


FIGURE 2 - A computer simulation of the time evolution of the various species in the NO-ClO₂-Br₂-HI system. The time axis corresponds to distance along the flow channel

TABLE II

Reactions considered in the computer simulation of the production of HBr
rate constants are in molecular units

Production and loss of chlorine atoms	Rate constant	Reference
1. $\text{ClO}_2 + \text{NO} \rightarrow \text{ClO} + \text{NO}_2$	3.4×10^{-13}	[7]
2. $\text{ClO} + \text{NO} \rightarrow \text{Cl} + \text{NO}_2$	1.7×10^{-11}	[8]
3. $\text{Cl} + \text{ClO}_2 \rightarrow 2\text{ClO}$	5.9×10^{-11}	[7]
4. $\text{ClO} + \text{ClO} \rightarrow \text{Cl} + \text{ClOO}$	2.8×10^{-14}	[9]
5. $\text{ClOO} + \text{M} \rightarrow \text{Cl} + \text{O}_2 + \text{M}$	1.34×10^{-14}	[2]
6. $\text{Cl} + \text{O}_2 + \text{M} \rightarrow \text{ClOO} + \text{M}$	5.6×10^{-34}	[10]
7. $\text{Cl} + \text{ClOO} \rightarrow \text{Cl}_2 + \text{O}_2$	1.56×10^{-10}	[11]
8. $\text{Cl} + \text{ClOO} \rightarrow 2\text{ClO}$	1.04×10^{-11}	[2]
9. $\text{ClO} + \text{ClO} + \text{M} \rightarrow \text{Cl}_2 + \text{O}_2 + \text{M}$	6.6×10^{-32}	[11]
10. $\text{Cl} + \text{Cl} + \text{M} \rightarrow \text{Cl}_2 + \text{M}$	6.39×10^{-33}	[12]
11. $\text{NO} + \text{Cl} + \text{M} \rightarrow \text{NOCl} + \text{M}$	9.3×10^{-32}	[13]
12. $\text{NOCl} + \text{Cl} \rightarrow \text{NO} + \text{Cl}_2$	3.0×10^{-11}	[14]
13. $\text{NO}_2 + \text{Cl} + \text{M} \rightarrow \text{NO}_2\text{Cl} + \text{M}$	7.2×10^{-31}	[15]
14. $\text{NO}_2\text{Cl} + \text{Cl} \rightarrow \text{Cl}_2 + \text{NO}_2$	3.0×10^{-11}	estimated
15. $\text{NO}_2 + \text{ClO} + \text{M} \rightarrow \text{NO}_3\text{Cl} + \text{M}$	1.0×10^{-31}	[16]
16. $\text{NO}_3\text{Cl} + \text{Cl} \rightarrow \text{NO}_2\text{Cl} + \text{ClO}$	2.2×10^{-12}	[17]
Production and loss of bromine atoms		
17. $\text{Cl} + \text{Br}_2 \rightarrow \text{BrCl} + \text{Br}$	1.2×10^{-10}	[4]
18. $\text{Cl} + \text{BrCl} \rightarrow \text{Br} + \text{Cl}_2$	1.45×10^{-11}	[4]
19. $\text{Br} + \text{Br} + \text{M} \rightarrow \text{Br}_2 + \text{M}$	3.35×10^{-33}	[12]
20. $\text{Br} + \text{Cl} + \text{M} \rightarrow \text{BrCl} + \text{M}$	4.5×10^{-33}	estimated
21. $\text{Br} + \text{NO} + \text{M} \rightarrow \text{NOBr} + \text{M}$	9.37×10^{-33}	[18]
22. $\text{Br} + \text{NOBr} \rightarrow \text{Br}_2 + \text{NO}$	3.65×10^{-10}	[18]
Laser pumping reaction		
23. $\text{Br} + \text{HI} \rightarrow \text{HBr} (\nu < 2) + \text{I}$	1.0×10^{-11}	[5]
Other reactions		
24. $\text{Cl} + \text{HI} \rightarrow \text{HCl} + \text{I}$	1.64×10^{-10}	[19]
25. $\text{I} + \text{I} + \text{M} \rightarrow \text{I}_2 + \text{M}$	6.67×10^{-33}	[20]

The initial concentration ratios of $[\text{NO}]:[\text{ClO}_2]:[\text{Br}_2]:[\text{HI}]$ were 2:1:1:1 with $[\text{ClO}_2]_0 = 1.0 \times 10^{15}$ molecules cm^{-3} . The NO/ClO_2 reaction produced the maximum Cl atom concentration at 750 μs at which time instantaneous injection of molecular bromine was simulated. Bromine atoms were formed rapidly with the bromine atom concentration reaching a maximum of 8.3×10^{14} molecules cm^{-3} within 125 μs of the addition of Br_2 . Hydrogen iodide addition was then simulated to form HBr . The rate of formation of HBr was sufficiently rapid to allow formation of 80 % of the maximum possible HBr concentration within the first 285 μs after injection of HI .

In this simulation, total HBr concentration rather than the concentration in individual vibrational levels of HBr was calculated. This was done because sufficient rate data does not exist to accurately describe the vibrational-vibrational (v-v) energy transfer reactions of HBr , the v-v energy transfer reactions of the HBr-HI systems and all vibrational-translational (v-t) processes which could effect $\text{HBr}(v=n)$. The dominant v-t deactivation process of $\text{HBr}(v=n)$ appears to be the deactivation by bromine atoms which has a rate constant of 2.6×10^{-12} $\text{cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ for $\text{HBr}(v=1)$ [21]. This is not surprising as the corresponding deactivation of $\text{HCl}(v=n)$ by chlorine atoms is an important loss process in HCl chemical lasers.

4.0 RESULTS AND DISCUSSION

Purely chemical HBr laser action at 4.0-4.1 μm was achieved in the present transverse-flow device. The optimum performance characteristics of the laser are given in Table III.

TABLE IIIHBr laser performance

Helium (sccm)	121,600
NO (sccm)	4,500
C ₂ O ₂ (sccm)	2,000
HI (sccm)	1,500
Br ₂ (sccm)	1,400
Total pressure (torr)	3.5
Wavelength (μm)	4.0-4.1
Total power (mW)	575

The total HBr multiline laser output power was measured as function of distance from the HI injector. As is shown in Fig. 3, a maximum power of 575 mW was achieved at a distance of 1 cm from the HI injector. This is $\sim 1/5$ of the total output power achieved under the same experimental conditions when the laser was operated as a HCl laser.

Lasing from HBr was observed on $v=1 \rightarrow 0$ transitions only with the $P_{1=0}(5)$ line being the most intense $\sim 75.5\%$, $P_{1=0}(6) \sim 21.9\%$ and $P_{1=0}(4) \sim 2.6\%$. Although lasing was observed on $v=2 \rightarrow 1$ transitions from the Br₂ - HI system by Chen (3), no lasing was observed from these transitions in the present system. This may be due to the fact that the most likely $v=2 \rightarrow 1$ transitions of HBr are strongly absorbed by CO₂. In the present device, external mirrors were used making effective flushing of the complete optical path difficult. Consequently the possibility of residual absorption by CO₂ cannot be ruled out.

Several improvements in the design of the HBr laser, which should result in considerably higher total output power, can be envisioned. Replacement of the rudimentary Br_2 injector by one similar to that used for the injection of HI and NO, but made of Teflon, should improve the mixing between Br_2 and the mainstream gas flow. The use of internal mirrors should eliminate the possibility of intra-cavity absorption of some of the HBr $v=2 \rightarrow 1$ transitions by CO_2 .

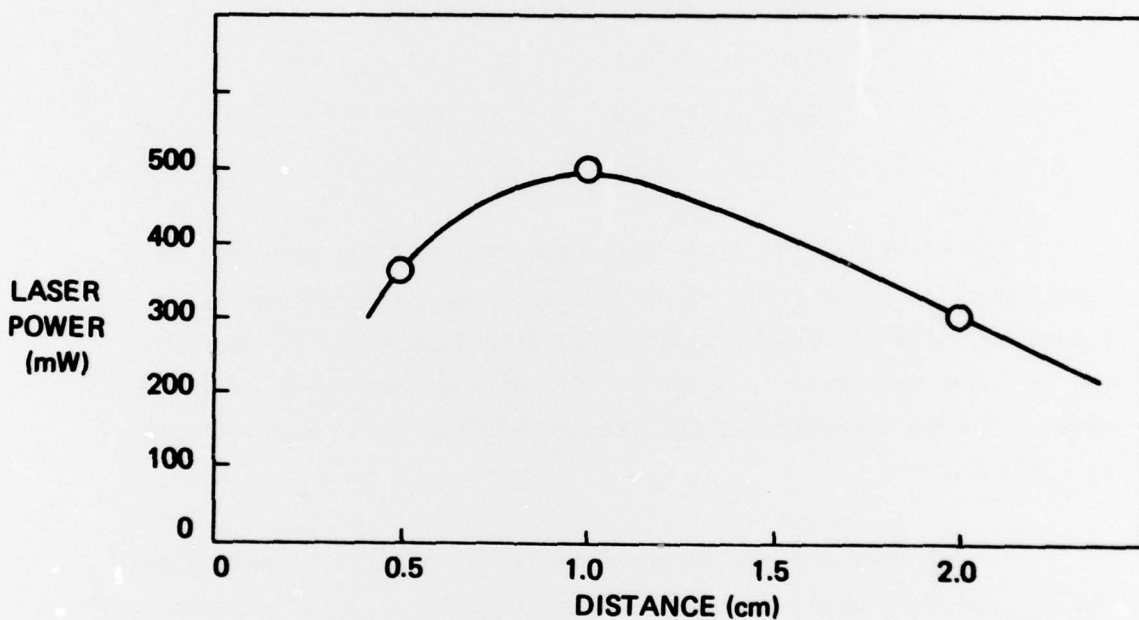


FIGURE 3 - Maximum multiline HBr output power as a function of distance from the HI injector

5.0 CONCLUSIONS

A purely chemical continuous wave HBr laser has been operated for the first time. Chlorine atoms, produced by the reaction of NO with ClO_2 in a transverse-flow system, react with molecular bromine forming bromine atoms which in turn react with HI to form vibrationally excited HBr.

Maximum HBr multiline total output powers of 575 mW have been obtained. Laser action was observed on three lines from the $v=1 \rightarrow 0$ vibrational band, $P_{1-0}(5)$, $P_{1-0}(6)$ and $P_{1-0}(4)$.

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Purely chemical laser action in HBr has been achieved for the first time. Chlorine atoms produced by the reaction of NO with ClO_2 were reacted with molecular bromine to form bromine atoms which in turn reacted with HI to form vibrationally excited HBr. Multiline HBr laser output of 0.58 W originated from the $P_{1-0}(4)$, $P_{1-0}(5)$ and $P_{1-0}(6)$ lines of the $v=1-0$ vibrational band. (U)

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